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DIFMIG - A COMPUTER PROGRAM FOR CALCULATION OF DIFFUSIVE
MIGRATION THROUGH MULTI-BARRIER SYSTEMS

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Abstract. The FORTRAN IV program DIFMIG calculates one-dimensionally (i.e. column) the diffusive migration of single substances through arbitrary multi-barrier systems according to the diffusion equation

$$\partial C / \partial t = D'(t) \partial^2 C / \partial x^2 + F'(C, t)$$

where $D'(t)$ is the effective dispersion coefficient and $F'(C, t)$ is a function responsible for time dependent changes in concentration other than dispersion/diffusion (e.g. slow dissolution of a compound from a repository, radioactive decay, and/or build up of daughter products. The method takes possible time dependent variations in the effective dispersion into account.

(continue on next page)

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The diffusion equation is solved by a finite difference implicit method, the resulting trigonal matrix equation being solved by standard methods.

INIS descriptors: D CODES; DIFFUSION LENGTH; FINITE DIFFERENCE METHOD; MIGRATION LENGTH; ONE-DIMENSIONAL CALCULATIONS; RADIO-NUCLIDE MIGRATION.

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1. INTRODUCTION

In recent years a series of computer codes, developed to study the migration behaviour of radionuclides through geological media, have appeared.¹ In general the aim of these programmes has been to predict the movement of radionuclides, following leakage from a repository, whereas minor attention has been drawn to the actual behaviour of the nuclides within the repository. Since the radioactive waste preferentially is, or will be, placed in repositories, consisting of several artificial barriers of very low permeability, it is expectable that ground water movements within the boundaries of the repository are negligible, i.e. any transport of dissolved material can be assumed to proceed by pure diffusion.

The computer program DIFMIG (which is written in FORTRAN IV) calculates, one-dimensionally (i.e. column), the diffusive migration of single substances through arbitrary multi-barrier systems, according to the diffusion equation (1)

$$\partial C / \partial t = D'(t) \partial^2 C / \partial x^2 + F'(C, t) \quad (1)$$

where $D'(t)$ is the effective dispersion coefficient and $F'(C, t)$ is a function responsible for the time dependent changes in concentration other than dispersion/diffusion, e.g. slow dissolution or leaching of a compound from a repository, radioactive decay, and/or build up of daughter products. The method furthermore, through $D'(t)$, takes the possible time dependent variations in the effective dispersion coefficient into account, the latter being defined as the product of the possibly time dependent retention factor $R_f(t)$ (2), and the diffusion coefficient in the liquid phase, D , (3). The total time dependence of the retention factor incorporates possible time dependences of the porosity, $\epsilon(t)$, as well as of the distribution coefficient $K_D(t)$. ρ is the density of the solid phase.

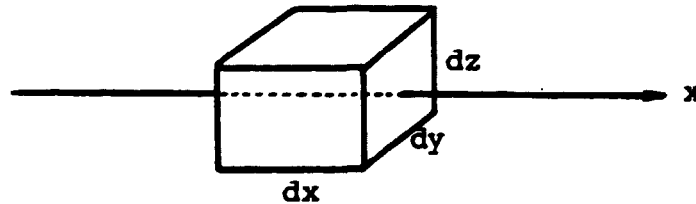
$$R_f(t) = (1 + \rho K_D(t) [1 - \epsilon(t)] / \epsilon(t))^{-1} \quad (2)$$

$$D'(t) = R_f(t) \cdot D \quad (3)$$

The diffusion equation (1) is solved by a finite difference implicit method, the resulting trigonal matrix equation being solved by standard methods.

2. SET UP THE DIFFUSION EQUATION

To set up the equation for the diffusive transport through a column, we shall look at the mass-balance for a single volume element $dx dy dz$ in the column



Assuming the flow of the component to be in the liquid phase only and only in the x -direction (i.e. the one-dimensional case) the mass-balance for a dissolved compound (e.g. a radionuclide) in the volume element $dx dy dz$ is given by the equation (4)

$$\begin{aligned} \epsilon dx dy dz \partial C / \partial t + \rho (1 - \epsilon) dx dy dz \partial \bar{C} / \partial t = & - \epsilon dy dz D (\partial C / \partial x)_x \\ & + \epsilon dy dz D (\partial C / \partial x)_{x+dx} + F(C, t) dx dy dz - \epsilon dx dy dz \lambda C \\ & - \rho (1 - \epsilon) dx dy dz \lambda \bar{C} \end{aligned} \quad (4)$$

where ϵ is the porosity, $J = -D \partial C / \partial x$ is the flux pr. unit area of liquid phase in the cross section $dy dz$ (the liquid flow area is $\epsilon dy dz$), D is the diffusion coefficient in the liquid phase, $F(C, t)$ is the total amount of the compound produced in a unit volume of the column per unit time at the time t , λ is the decay constant of the radionuclide, ρ is the density of the solid phase, and C and \bar{C} are the concentrations of the component in the

liquid phase and on the solid phase, respectively, the latter being equal to $K_D C$, where K_D is the distribution coefficient.

By introducing $R_f = (1 + \rho K_D (1 - \epsilon) / \epsilon)^{-1}$ and by expanding $(\partial C / \partial x)_{x+dx}$ in Taylor series the mass-balance are reduced to equation (5), assuming equilibrium sorption of the radionuclide, an assumption which in general will be valid, since the equilibrium sorption will be rapid compared to the possible time dependent changes in retention factors and porosities.

$$\partial C / \partial t = R_f D \partial^2 C / \partial x^2 + R_f F(C, t) / \epsilon - \lambda C \quad (5)$$

which by introduction of equation (3) as well as the time dependence of R_f and ϵ (cf. (2)) leads to equation (1) adopting (6)

$$F'(C, t) = R_f(t) F(C, t) / \epsilon(t) - \lambda C \quad (6)$$

3. SOLUTION OF THE DIFFUSION EQUATION

The finite difference representations of the single elements in the diffusion equation (1) is given by the equations (7) through (9), designating the indices j and i to time and distance, respectively.

$$(\partial C / \partial t)_{i,j} = (C_{i,j+1} - C_{i,j}) / \Delta j \quad (7)$$

$$(\partial C / \partial x)_{i,j} = (C_{i+1,j} - C_{i-1,j}) / 2\Delta i \quad (8)$$

$$(\partial^2 C / \partial x^2)_{i,j} = (C_{i+1,j} + C_{i-1,j} - 2C_{i,j}) / \Delta i^2 \quad (9)$$

Accordingly, the diffusion equation is represented by backward differences in time for implicit solution (10)

$$\begin{aligned} (C_{i,j+1} - C_{i,j}) / \Delta j = D'_i(t_{j+1}) (C_{i+1,j+1} + C_{i-1,j+1} - 2C_{i,j+1}) / \Delta i^2 \\ + F'(C_{i,j+1}, t_{j+1}) \end{aligned} \quad (10)$$

For functions $F'(C, t)$, which vary only slowly with time, the latter term in equation (10) is approximated by $F'(C_{i,j}, t_j)$ in order to preserve the trigonal system of equations. The general equation to be solved, (11), is then set up by rewriting (10)

$$-A_i C_{i-1,j+1} + (1+2A_i) C_{i,j+1} - A_i C_{i+1,j+1} = C_{i,j} + \Delta j F'(C_{i,j}, t_j) \quad (11)$$

where $A_i = D'(t_{j+1}) \Delta j / \Delta i^2$

The general equation (11) is valid for all points through out the column except for a) the first point in the column (i.e. $i = 0$), b) points where properties (e.g. D' and ϵ) change abruptly (i.e. boundaries between the single barriers), and c) the end of the column. These three cases are to be handled separately in the following.

The arrangement of the repository and the surrounding barriers is visualized schematically in figure I.

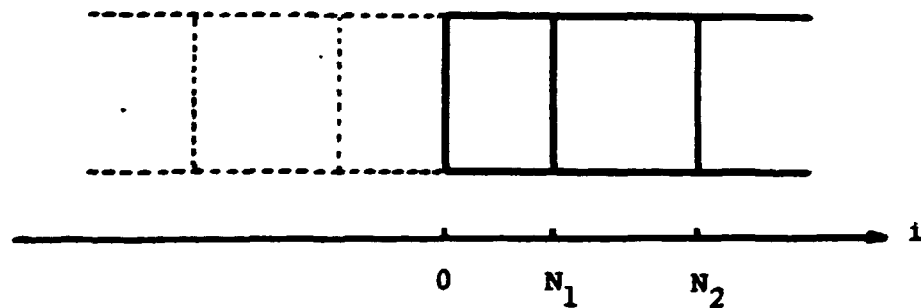


Figure I. Schematically representation of the repository and the surrounding barriers. For calculational reasons (vide infra) a symmetrical arrangement of the barriers around the center of the repository is assumed.

3.1 First Point in the Column

Assuming a symmetrical arrangement of the repository and the surrounding barriers (cf. fig. I), the center of the repository appears as the first point in the column ($i = 0$). The boundary condition for this point is given by equation (12)

$$(\partial C / \partial x)_{i=0} = (C_{i=1} - C_{i=-1}) / 2\Delta i = 0 \quad (12)$$

The equation for the first point in the column, equation (13), is accordingly obtained by insertion of (12) into the general equation (11)

$$i=0: \quad (1+2A_0)C_{0,j+1} - 2A_{0,j+1}C_{1,j+1} = C_{0,j} + \Delta j F'(C_{0,j}, t_j) \quad (13)$$

3.2 Boundaries Between Barriers

When the barrier properties (D' and ϵ) changes abruptly at points $i = N_K$ (cf. fig. I), the boundary condition for these points are given by equation (14)

$$\epsilon_K J_{N_K}^K = \epsilon_{K+1} J_{N_K}^{K+1} \quad (14)$$

where K is the barrier number and $i=N_K$ is the point where the barriers K and $K+1$ meets (cf. fig. I).

In order to avoid an introduction of a relation between two fictive concentrations, $C_{N_K+1}^K$ and $C_{N_K-1}^{K+1}$, and to preserve the trigonal system of equations, the concentration gradients in the expressions for $J_{N_K}^K$ and $J_{N_K}^{K+1}$ are represented by simple forward differences instead of central differences (equation (15)).

$$\epsilon_K J_{N_K}^K = -D_{N_K}^K \epsilon_K (C_{N_K} - C_{N_K-1}) / \Delta i = \epsilon_{K+1} J_{N_K}^{K+1} = -D_{N_K}^{K+1} \epsilon_{K+1} (C_{N_K+1} - C_{N_K}) / \Delta i \quad (15)$$

which gives

$$C_{N_K} = E_K D_{N_K}^K C_{N_K-1} / (E_K D_{N_K}^K + D_{N_K}^{K+1}) + D_{N_K}^{K+1} C_{N_K+1} / (E_K D_{N_K}^K + D_{N_K}^{K+1}) \quad (16)$$

where

$$E_K = \epsilon_K / \epsilon_{K+1} \quad (17)$$

Hence, the calculation of C_{N_K} is not a part of the integration procedure. Accordingly, the integration equations for the last

point in the barrier K and the first point in the barrier K+1 are given by the equations (18) and (19), respectively.

$$-A_{N_K-1}C_{N_K-2,j+1} + (1+2A_{N_K-1}-\delta_{N_K}A_{N_K-1})C_{N_K-1,j+1} - (1-\delta_{N_K})A_{N_K-1}C_{N_K+1,j+1} = C_{N_K-1,j} + \Delta j F'(C_{N_K-1,j}, t_j) \quad (18)$$

$$-\delta_{N_K}A_{N_K+1}C_{N_K-1,j+1} + (1+2A_{N_K+1}-(1-\delta_{N_K})A_{N_K+1})C_{N_K+1,j+1} - A_{N_K+1}C_{N_K+2,j+1} = C_{N_K+1,j} + \Delta j F'(C_{N_K+1,j}, t_j) \quad (19)$$

where

$$\delta_{N_K} = E_K D_{N_K}^K / (E_K D_{N_K}^K + D_{N_K+1}^{K+1}) \quad (20)$$

3.3 Boundary Condition at the End of the Last Barrier.

Two different types of barrier systems are considered: a) the last barrier is of infinite length, and b) at the point $i=N_M$ the last barrier ends in a pond.

3.3.1 Last barrier of infinite length.

Assuming the last barrier being of infinite length, the calculation stops at a point $i=N_M$, which should be a point in the column that is never reached by the progressing concentration profile during the time period covered by the calculation.

At this point

$$(\partial C / \partial x)_{N_M} = (C_{N_M+1} - C_{N_M-1}) / 2\Delta i = 0 \quad (21)$$

which leads to the following last integration equation to be solved (22)

$$-2A_{N_M}C_{N_M-1,j+1} + (1+2A_{N_M})C_{N_M,j+1} = C_{N_M,j} + \Delta j F'(C_{N_M,j}, t_j) \quad (22)$$

3.3.2 Last barrier ends in a pond

Assuming a) the last barrier at a point $i=N_M$ to end in a pond of volume = V, b) the water within the pond to be completely mixed at all times (i.e. the concentration in the water, C_W , is constant through out the pond), c) that a constant flow of water, WP , is withdrawn from the pond, and d) the flow of the migrating species across the boundary layer between the barrier and the pond to be stationary and free of simple convection in the direction of transport at all times, the boundary condition at the end of the final barrier can be approximated by equation (23)

$$\epsilon_{NB} J_{N_M} = -D_{N_M} \epsilon_{NB} (C_{N_M+1} - C_{N_M-1}) / 2\Delta i = -(C_W - C_{N_M}) D_W / \delta_W \quad (23)$$

which may be rewritten into equation (24)

$$C_{N_M+1} = C_{N_M-1} - (2\Delta i / \epsilon_{NB} D_{N_M}) (D_W / \delta_W) C_{N_M} + (2\Delta i / \epsilon_{NB} D_{N_M}) (D_W / \delta_W) C_W \quad (24)$$

where the subscript NB refers to the number of the final barrier and D_W / δ_W is the mass transfer number for the boundary layer.

Combining the equations (24) and (11) affords the last integration equation in the last barrier (25)

$$-2A_{N_M} C_{N_M-1,j+1} + (1+2A_{N_M} + A_{N_M} (2\Delta i / \epsilon_{NB} D_{N_M}) (D_W / \delta_W)) C_{N_M,j+1} - A_{N_M} (2\Delta i / \epsilon_{NB} D_{N_M}) (D_W / \delta_W) C_W = C_{N_M,j} + \Delta j F'(C_{N_M,j}, t_j) \quad (25)$$

When the bulk area of the column through which the migrating species is transferred from the last barrier into the pond is A, equation (26) is valid

$$d(C_W V) / dt = -WPC_W + A \epsilon_{NB} J_{N_M} - \lambda C_W V \quad (26)$$

which for constant volume, V, and by introduction of (23) can be rewritten into (27)

$$dC_W / dt = -WPC_W / V - (A(C_W - C_{N_M}) / V) D_W / \delta_W - \lambda C_W \quad (27)$$

Representing this equation (27) by its finite difference approximation, of the same type as used for the single barriers, we obtain equation (28), which is the last equation to be integrated.

$$-(\Delta j A/V)(D_W/\delta_W)C_{N_M,j+1} + (1+\Delta j W F/V+(\Delta j A/V)(D_W/\delta_W))C_{W,j+1} = C_{W,j} - \lambda C_W \Delta j \quad (28)$$

The equations (11), (13), (18), (19), and (22) (or (25) and (28)) enable us to set up the total trigonal system of equations, which is solved by standard methods.²

4. REFERENCES

- 1) see e.g. H.C. Burkholder and E.L.J. Rosinger, "A model for the transport of radionuclides and their decay products through geologic media" AECL-6325, Whiteshell Nuclear Research Establishment, 1979; W.V. DeMier, M.O. Cloninger, H.C. Burkholder, and P.J. Liddell, "GETOUT-A computer program for predicting radionuclide decay chain transport through geologic media" PNL-2970, Pacific Northwest Laboratory, 1979; K.L. Kipp, "Mathematical modelling of chemical transport in soil columns" AERE-R 9012, Harwell, 1978; J. Hadermann, Nucl. Technol. 47 312 (1980)
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- 8) J. Crank, "The mathematics of diffusion" Clarendon Press, Oxford, 1975, p.15

APPENDIX I: COMPUTER CODE

```

100 $RESET FREE
200 FILE 7(TITLE='MIGRAC',KIND=DISK,MAXRECSIZE=22)
300 FILE 8(TITLE='MIGRAF',KIND=DISK,MAXRECSIZE=22)
400 FILE 9(TITLE='MIGRAW',KIND=DISK,MAXRECSIZE=22)
500 C THE PROGRAM "DIFMIG" CALCULATES ONE-DIMENTIONALLY (I.E. COLUMN)
600 C THE DIFFUSIVE MIGRATION OF SINGLE SUBSTANCES THROUGH A MULTI-
700 C BARRIER SYSTEM, ACCORDING TO THE DIFFUSION EQUATION:
800 C  $(\partial C/\partial T) = D^* (\partial^2 C/\partial X^2) + F(C,T)$ 
900 C WHERE D* IS THE EFFECTIVE DISPERSION COEFFICIENT AND F(C,T) IS
1000 C A FUNCTION RESPONSIBLE FOR TIME DEPENDENT CHANGES IN CONCENTRA-
1100 C TION OTHER THAN DISPERSION/DIFFUSION (E.G. SLOW DISSOLUTION
1200 C OF THE COMPOUND FROM A REPOSITORY AND/OR RADIOACTIVE DECAY.
1300 C THE METHOD FURTHERMORE TAKES A POSSIBLE TIME DEPENDENT VARIATION
1400 C IN THE EFFECTIVE DISPERSION COEFFICIENTS INTO ACCOUNT.
1500 C THE DIFFUSION EQUATION IS SOLVED BY A FINITE DIFFERENCE IM-
1600 C PLOCITE METHOD; THE RESULTING TRIGONAL MATRIX EQUATION IS
1700 C SOLVED BY STANDARD METHODS.
1800 C REFERENCE: P.BØ AND L.CARLSEN, RISØE-M-2262, 1981.
1900 C THE FOLLOWING PARAMETERS ARE NEEDED
2000 C "NB" : NUMBER OF BARRIERS
2100 C "NMAX" : NUMBER OF THE LAST POINT IN THE COLUMN
2200 C "DELT" : THE CONSTANT TIME STEP USED IN THE CALCULATION
2300 C "DELN" : THE EQUIDISTANT DISTANCE BETWEEN THE POINTS THROUGH
2400 C THE COLUMN
2500 C "RECIP" : AN INTEGER INDICATING THE TYPE OF 'END BOUNDARY
2600 C CONDITION'. '0' INDICATES AN INFINITE FINAL
2700 C BARRIER; '1' INDICATES OUTFLOW INTO A WELL
2800 C "DW" : MASS TRANSPORT FIGURE FOR WATER (USED IN THE CASE
2900 C OF OUTFLOW INTO A WELL). "DW" IS THE RATIO BETWEEN
3000 C THE DIFFUSION COEFFICIENT IN WATER AND THE THICK-
3100 C NESS OF THE BOUNDARY LAYER
3200 C "AREA" : COLUMN CROSS SECTION AREA (BULK AREA)
3300 C "VOL" : VOLUME OF THE WELL
3400 C "WF" : THE CONTINUOUS PUMP OUT FROM THE WELL
3500 C "DECAY" : RADIOACTIVE DECAY CONSTANT
3600 C "H0","H1" : CONSTANTS IN THE F(C,T) FUNCTION
3700 C "TMAX" : MAXIMUM TIME FOR THE MIGRATION STUDY
3800 C "TOPL" : MAXIMUM TIME FOR THE POSSIBLE DISSOLUTION OF MA-
3900 C TERIAL FROM THE REPOSITORY. IN GENERAL BARRIER-1
4000 C WILL BE DESIGNATED TO THE REPOSITORY
4100 C "KCT" : NUMBER OF TIMESTEPS BETWEEN OUTPUT OF CONCENTRATION PRO
4200 C FILE
4300 C "KFT" : NUMBER OF TIMESTEPS BETWEEN COLLECTION OF OUTFLOW THROU
4400 C GH THE SINGLE BARRIER BOUNDARIES
4500 C "KRT" : NUMBER OF TIMESTEPS BETWEEN CHANGE IN RETENTION FACTORS
4600 C "KET" : NUMBER OF TIMESTEPS BETWEEN CHANGE IN POROSITIES
4700 C CIENTS (AS A FUNCTION OF TIME)
4800 C DIMENSION C(2,1000),NDB(10),F(1000),D(1000),RF0(1000),RFF(1000),GF
4900 C 1(1000),RF(1000),EPS0(1000),EPSF(1000),GEPS(1000),EPS(1000),A(1000)
5000 C 2,A1(1000),A2(1000),A3(1000),ALPHA(1000),BETA(1000),Y(1000),CU(1000
5100 C 3),FL(10,2000),CW(2000)

```

```

5200      INTEGER RECIP
5300      READ(5,/) NB,NMAX,DELT,DELN,RECIP,DW,AREA,VOL,UF,DECAY,H0,H1,TMAX,
5400      ITOPL
5500      READ(5,/) KCT,KFT,KRT,KET
5600 C      "NBB(I)" IS THE LAST POINT FOR CALCULATION IN THE BARRIER I
5700      DO 200 I=1,NB
5800      READ(5,/) NBB(I)
5900 200    CONTINUE
6000      T=0
6100      IC3=1
6200      IC1=2
6300      IC2=1
6400 C      "T" IS THE TIME AND IC3, IC1, AND IC2 ARE CONCENTRATION
6500 C      ARRAY PARAMETERS
6600      KC=0
6700      KF=0
6800      KKF=0
6900 C      "KC" AND "KF" COUNT FROM 0 TO KCT AND KFT, RESPECTIVELY
7000 C      "KKF" COUNTS THE NUMBER OF KFT INTERVALS (EQUAL TO (TMAX/DELT)+1)
7100      DO 220 I=1,NMAX+1
7200      F(I)=-DECAY
7300 C      THE "F(I)" FUNCTION IS RESPONSIBLE FOR CHANGES IN CONCENTRATION
7400 C      DUE TO POSSIBLE RADIOACTIVE DECAY
7500 220    CONTINUE
7600 C**** INITIAL CONCENTRATION PROFILE ****
7700 C      "C0,N1,N2" CORRESPONDS TO THE CONCENTRATION C AT THE TIME=0
7800 C      IN THE COLUMN SECTION LIMITED BY THE POINTS N1 AND N2
7900 310    READ(5,/) Q,N1,N2
8000      DO 300 I=N1,N2,1
8100      C(I,1)=Q
8200 300    CONTINUE
8300      IF (N2.LT.NMAX) GOTO 310
8400 C**** DISPERSION COEFFICIENTS (INPUT)****
8500 C      THE DISPERSION COEFFICIENTS ("D(I)") INCORPORATES EFFECTS OF
8600 C      DIFFUSION AND DISPERSION
8700 C      "D,N1,N2" CORRESPONDS TO THE DISPERSION COEFFICIENTS WITHIN
8800 C      THE COLUMN RANGE LIMITED BY THE POINTS N1 AND N2
8900 420    READ(5,/) Q,N1,N2
9000      DO 410 I=N1,N2,1
9100      D(I)=Q
9200 410    CONTINUE
9300      IF (N2.LT.NMAX) GOTO 420
9400 C      "RFO" AND "RFF" ARE THE RETENTION FACTORS AT THE TIME=0 AND TIME=I
9500 C      INFINITE, RESPECTIVELY. "GR" IS A CONSTANT.
9600 C      'RFO' AND 'RFF' IN THE COLUMN SECTION LIMITED BY THE POINTS N1 AND
9700 C      N2
9800 440    READ(5,/) R,S,Q,N1,N2
9900      DO 430 I=N1,N2,1
10000     RFO(I)=R
10100     RFF(I)=S
10200     GR(I)=Q

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10300 430  CONTINUE
10400      IF(N2.LT.NMAX) GOTO 440
10500 C      "EPS0" AND "EP3F" ARE THE POROSITIES AT THE TIME=0 AND TIME=INFINI
10600 C      TE, RESPECTIVELY. "GEPs" IS A CONSTANT.
10700 C      'EPS0' AND 'EPSF' IN THE COLUMN SECTION LIMITED BY THE POINTS
10800 C      N1 AND N2
10900 460  READ(5,/)R,S,Q,N1,N2
11000      DO 450 I=N1,N2,1
11100      EPS0(I)=R
11200      EPSF(I)=S
11300      GEPs(I)=Q
11400 450  CONTINUE
11500      IF(N2.LT.NMAX) GOTO 460
11600 C**** PLOTTER PARAMETERS FOR OUTPUT FILES MIGRAC ,MIGRAF, AND MIGRAU****
11700 C      THIS SECTION IN THE PROGRAM MAY BE SUBSTITUTED SUITABLY IF
11800 C      ANOTHER PLOTTING PROCEDURE FOR THE OUTPUT FILES IS PREFERRED.
11900      READ(5,/) RMAX,SMAX
12000      R=NMAX/RMAX
12100      S=TMAX/(KFT*DELT*SMAX)
12200 C      "RMAX", "SMAX", "R", AND "S" ARE USED FOR THE POSSIBLE PLOTTING
12300 C      OF THE OUTPUT FILES (MIGRAC/MIGRAF/MIGRAU) BY THE PROGRAM MIGPLOT
12400 C      "RMAX" AND "SMAX" ARE THE TOTAL MIGRATION PATHWAY AND TIME, RESPEC
12500 C      TIVELY IN UNITS SUITABLE FOR THE PLOTTER ROUTINE.
12600 C      OBS!!!! RMAX AND SMAX MUST BE LESS THAN 10, E.G. FOR PATHWAYS UP
12700 C      TO 100 METERS AND TIMES UP TO 100 YEARS LMAX AND NMAX MUST BE GIVE
12800 C      N IN DECAMETERS AND DECAYEARS, RESPECTIVELY. FOR PATHWAYS AND TIME
12900 C      S UP TO 1000 METERS AND YEARS IN HEKTOMETERS AND HEKTOYEARS
13000 C      RESPECTIVELY ETC.
13100 C**** CALCULATION OF EFFECTIVE DISPERSION COEFFICIENTS, RETENTION
13200 C      FACTORS AND POROSITIES
13300 C      "RF(I)" IS THE RETENTION FACTORS TO THE TIME=T
13400 C      "EPS(I)" IS THE POROSITIES TO THE TIME=T
13500      KR=0
13600      KE=0
13700 C      "KR" AND "KE" COUNTS FROM 0 TO KRT AND KET, RESPECTIVELY
13800 830  DO 470 I=1,NMAX
13900      RF(I)=RFF(I)-(RFF(I)-RFO(I))*EXP(-GR(I)*T)
14000      Q=RF(I)*D(I)
14100      A(I)=DELT*Q/DELN**2
14200 470  CONTINUE
14300      KR=0
14400      IF(T.EQ.0) GOTO 840
14500      IF(KE.LT.KET) GOTO 490
14600 840  DO 480 I=1,NMAX
14700      EPS(I)=EPSF(I)-(EPSF(I)-EPS0(I))*EXP(-GEPs(I)*T)
14800 480  CONTINUE
14900      KE=0
15000 490  CONTINUE
15100 C**** TRIGONAL MATRIX EQUATION COEFFICIENTS****
15200 C      "A1(I)", "A2(I)", "A3(I)" ARE THE TRIGONAL MATRIX COEFFICIENTS
15300      A1(I)=0.

```

```
15400      A2(1)=1.+2.*A(1)
15500      A3(1)=-2.*A(1)
15600      DO 500 I=2,NBB(1)-1
15700      A1(I)=-A(I)
15800      A2(I)=1.+2.*A(I)
15900      A3(I)=-A(I)
16000 500   CONTINUE
16100      DO 510 I=2,NB
16200      K=NBB(I-1)
16300      Q=EPS(K)*D(K)/EPS(K+1)
16400      DELG=Q/(Q+D(K+1))
16500      A1(K)=-A(K)
16600      A2(K)=1.+2.*A(K)-DELG*A(K)
16700      A3(K)=- (1.-DELG)*A(K)
16800      A1(K+1)=-DELG*A(K+1)
16900      A2(K+1)=1.+2.*A(K+1)- (1-DELG)*A(K+1)
17000      A3(K+1)=-A(K+1)
17100      DO 520 J=K+2,NBB(I)-1
17200      A1(J)=-A(J)
17300      A2(J)=1.+2.*A(J)
17400      A3(J)=-A(J)
17500 520   CONTINUE
17600 510   CONTINUE
17700      A1(NMAX-1)=-A(NMAX-1)
17800      A2(NMAX-1)=1.+2.*A(NMAX-1)
17900      A3(NMAX-1)=-A(NMAX-1)
18000      IF(RECIP.EQ.1) GOTO 600
18100      A1(NMAX)=-2.*A(NMAX)
18200      A2(NMAX)=1.+2.*A(NMAX)
18300      A3(NMAX)=0.
18400      GOTO 700
18500 600   A1(NMAX)=-2.*A(NMAX)
18600      A2(NMAX)=1.+2.*A(NMAX)+2.*DELT*DW*RF(NMAX)/(DELN*EPS(NMAX))
18700      A3(NMAX)=-2.*DELT*DW*RF(NMAX)/(DELN*EPS(NMAX))
18800      A1(NMAX+1)=-DELT*AREA*DU/VOL
18900      A2(NMAX+1)=1.+DELT*UF/VOL + DELT*AREA*DU/VOL
19000      A3(NMAX+1)=0.
19100      GOTO 700
19200 700   CONTINUE
19300 C***  MATRIX COEFFICIENTS***
19400      NM=NMAX
19500      IF(RECIP.EQ.1) NM=NMAX+1
19600      ALPHA(1)=A2(1)
19700      BETA(1)=A3(1)/ALPHA(1)
19800      DO 710 I=2,NM
19900      ALPHA(I)=A2(I)-A1(I)*BETA(I-1)
20000      BETA(I)=A3(I)/ALPHA(I)
20100 710   CONTINUE
20200 C***  INTEGRATION***
20300 820   IC3=-IC3
20400      IC1=IC1+IC3
```

```

20500      IC2=IC2-IC3
20600      IF(T.GT.TOPL)H0=0.
20700      IF(T.GT.TOPL)H1=0.
20800      Y(I)=(C(IC1,I)+DELT*(F(I)*C(IC1,I)+(RF(I)/EPS(I))*H0*EXP(-H1*T)*EX
20900      1P(F(I)*T)))/ALPHA(I)
21000      DO 800 I=2,NM
21100      Q=F(I)*C(IC1,I)
21200      IF(I.LE.NBB(1))Q=Q+(RF(I)/EPS(I))*H0*EXP(-H1*T)*EXP(F(I)*T)
21300      Y(I)=(C(IC1,I)+DELT*Q - A1(I)*Y(I-1))/ALPHA(I)
21400 800  CONTINUE
21500      C(IC2,NM)=Y(NM)
21600      DO 810 I=1,NM-1
21700      K=NM-I
21800      C(IC2,K)=Y(K)-BETA(K)*C(IC2,K+1)
21900 C      THE ARRAY C(IC2,K) INCLUDES THE CONCENTRATIONS IN ALL POINTS
22000 C      IN THE COLUMN OTHER THAN THOSE DESIGNATED TO BARRIER BOUNDARIES
22100 C      NOTE: THE POINTS ARE CONTINUOUSLY NUMBERED
22200 810  CONTINUE
22300      KC=KC+1
22400      KF=KF+1
22500      KR=KR+1
22600      KE=KE+1
22700      T=T+DELT
22800 C**** RESULTS/OUTPUTFILES****
22900 C      OUTPUTFILE MIGRAC CONTAINS CONCENTRATION PROFILES (CI/M**3)
23000 C      OUTPUTFILE MIGRAF CONTAINS FLUXES OUT THROUGH THE SINGLE BARRIERS
23100 C      (CI/YEAR*M**2(BULK AREA))
23200 C      OUTPUTFILE MIGRAW CONTAINS CONCENTRATIONS IN WELL (CI/M**3)
23300      IF(KC.EQ.KCT) GOTO 1000
23400 900  CONTINUE
23500      IF(KF.EQ.KFT) GOTO 1100
23600 910  CONTINUE
23700      IF(KR.EQ.KRT) GOTO 830
23800      IF(KE.EQ.KET) GOTO 840
23900      IF(T.GT.TMAX) GOTO 1200
24000      GOTO 820
24100 1000 CONTINUE
24200      DO 1010 I=1,NBB(1),1
24300      CU(I)=C(IC2,I)
24400 C      THE ARRAY CU(I) INCLUDES CONCENTRATIONS IN ALL POINTS IN THE COLUM
24500 C      N, INCLUDING THE POINTS DESIGNATED TO THE BARRIER BOUNDARIES
24600 1010 CONTINUE
24700      I=NBB(1)+1
24800      Q=EPS(I-1)*D(I-1)/EPS(I)
24900      DELG=Q/(Q+D(I))
25000      CU(I)=DELG*C(IC2,I-1)+(1-DELG)*C(IC2,I)
25100      DO 1020 J=2,NB
25200      DO 1030 K=NBB(J-1)+1,NBB(J)
25300      CU(K+J-1)=C(IC2,K)
25400 1030 CONTINUE
25500      IF(J.EQ.NB) GOTO 1040

```

```

25600      Q=EPS(NBB(J))*D(NBB(J))/EPS(NBB(J)+1)
25700      DELG=Q/(Q+D(NBB(J)+1))
25800      CU(NBB(J)+J)=DELG*C(IC2,NBB(J))+(1-DELG)*C(IC2,NBB(J)+1)
25900 1020  CONTINUE
26000 1040  CU(NBB(NB)+NB)=C(IC2,NMAX)
26100      IF(RECIP.EQ.1) CU(NBB(NB)+NB+1)=C(IC2,NM)
26200      WRITE(7,1060) T
26300 1060  FORMAT(X,E10.3)
26400      WRITE(7,/) NM,0,R
26500      WRITE(7,1050) (CU(J),J=1,NM)
26600 1050  FORMAT(8E10.3)
26700      KC=0
26800      GOTO 900
26900 1100  CONTINUE
27000      KKF=KKF+1
27100      DO 1110 J=1,NB-1
27200      Q=EPS(NBB(J))*D(NBB(J))/EPS(NBB(J)+1)
27300      DELG=Q/(Q+D(NBB(J)+1))
27400      Q=DELG*C(IC2,NBB(J))+(1-DELG)*C(IC2,NBB(J)+1)
27500      FL(J,KKF)=-D(NBB(J))/DELN*(Q-C(IC2,NBB(J)))*EPS(NBB(J))
27600 C      THE 'FL' ARRAY COLLECTS THE FLUXES OUT THROUGH THE SINGLE BARRIERS
27700 1110  CONTINUE
27800      KF=0
27900      IF(RECIP.EQ.0) GOTO 910
28000      FL(NB,KKF)=-DW*(C(IC2,NM)-C(IC2,NM-1))
28100      CU(KKF)=C(IC2,NM)
28200 C      THE ARRAY CU COLLECTS THE CONCENTRATIONS IN THE WELL (CORRESPONDING
28300 C      1 TO RECIP=1)
28400      GOTO 910
28500 1200  CONTINUE
28600      CLOSE(7,DISP=KEEP)
28700      DO 1230 J=1,NB
28800      WRITE(8,1210) J
28900 1210  FORMAT(X,I3)
29000      WRITE(8,/) KKF,0,S
29100      WRITE(8,1220) (FL(J,I),I=1,KKF)
29200 1220  FORMAT(8E10.3)
29300 1230  CONTINUE
29400      CLOSE(8,DISP=KEEP)
29500      IF(RECIP.EQ.0) GOTO 1300
29600      WRITE(9,1310)
29700 1310  FORMAT(' WELL CONCENTRATION'/)
29800      WRITE(9,/) KKF,0,S
29900      WRITE(9,1320) (CU(J),J=1,KKF)
30000 1320  FORMAT(8E10.3)
30100      CLOSE(9,DISP=KEEP)
30200 1300  CONTINUE
30300      STOP
30400      END
#

```

APPENDIX II: POROSITIES

In table AII-1 the porosities for a series of common geological materials are given.

TABLE AII-1. Porosities of selected materials.³

material	porosity (ϵ) (in pct)
sand	37-50
granular crushed rock	44-45
soil	43-54
sand stone	3-38
limestone (dolomite)	4-10
coal	2-12
sandy clay	15-25
clay	40-50
chalk	30-50
concrete (ord. mix.)	2- 7
spherical packings	36-43

APPENDIX III: DIFFUSION COEFFICIENTS

In a porous media (without any chemical interaction between the diffusing species and the solid phase) the apparent diffusion coefficient, measured relative to the open ends of a tortuous pore, is less than the intrinsic diffusion coefficient in the pore fluid by a factor proportional to the square of the tortuosity of the pore, since the actual path is increased proportionally to the tortuosity and the concentration gradient along the pore is reduced proportionally to the tortuosity.

According to the theory of Mackie and Meares⁴ the apparent diffusion coefficient, D , is given by equation (AIII-1), where D_{aq} refers to the diffusion coefficient in pure water.

$$D = D_{aq} (\epsilon/2 - \epsilon)^2 \quad (\text{AIII-1})$$

However, it should be noted that equation (AIII-1) applies to homogeneous media and only when the pore diameter is large compared to the diameter of the diffusing molecule.

A distinct decreasing effect of the apparent diffusion coefficient with decreasing pore size has been observed,⁵ leading to a revised version of the equation for D (equation (AIII-2)).

$$D = D_{aq} Q (\epsilon/2 - \epsilon)^2 \quad (\text{AIII-2})$$

where Q is a constant less than 1. The actual magnitude of Q is dependent of the nature of the porous media and the diffusing material. In a recent investigation⁶ the factor Q has been determined to be ca. 0.05 for the diffusion of sodium ions in chalk samples from the Erslev chalk formation (Mors, Denmark).

The diffusion coefficients for ions, i , in pure water, D_{aq} , can be calculated by application of the limiting ionic conductances (λ_i ($\Omega^{-1}\text{cm}^2$)) in water (equation (AIII-3)).⁷

$$D_{aq}^i = \lambda_i RT / |z_i|^2 F^2 \quad (\text{AIII-3})$$

where R is the gas constant, T is the absolute temperature, z_i is the ionic charge and F is the Faraday.

Diffusion coefficients for selected ions are given in Table AIII-I

TABLE AIII-I. Diffusion coefficients ($\times 10^5 \text{ cm}^2 \text{ s}^{-1}$) for selected ions in pure water at 25°C

H^+ : 9.30	Mg^{2+} : 0.71	La^{3+} : 0.62	Cl^- : 2.03
Li^+ : 1.03	Ca^{2+} : 0.79	Ce^{3+} : 0.62	Br^- : 2.08
Na^+ : 1.33	Sr^{2+} : 0.79		I^- : 2.04
K^+ : 1.96	Ba^{2+} : 0.85		HCO_3^- : 1.45
Rb^+ : 2.07	Cu^{2+} : 0.72		SO_4^{2-} : 0.53
Cs^+ : 2.06	Zn^{2+} : 0.70		

APPENDIX IV: TIME DEPENDENCE OF POROSITIES AND RETENTION FACTORS

The program DIFMIG takes into account the possibilities of time dependent changes of porosities, $\epsilon_K(t)$, and retention factors, $R_f^K(t)$.

Since no mathematical formulations of possible time dependence of porosities and retention factors have been reported, the program DIFMIG operates with simple exponential changes between initial values, $\epsilon_K(t_0)$ and $R_f^K(t_0)$, to the time $t = t_0$, and final values, $\epsilon_K(t_\infty)$ and $R_f^K(t_\infty)$, reached at infinite time, $t = t_\infty$. Intermediary values of porosities and retention factors at times $t = t_j$ are given by the equations (AIV-1) and (AIV-2), G_ϵ and G_R being constants.

$$\epsilon_K(t_j) = \epsilon_K(t_\infty) - (\epsilon_K(t_\infty) - \epsilon_K(t_0)) \exp(-G_\epsilon t_j) \quad (\text{AIV-1})$$

$$R_f^K(t_j) = R_f^K(t_\infty) - (R_f^K(t_\infty) - R_f^K(t_0)) \exp(-G_R t_j) \quad (\text{AIV-2})$$

Other possible mathematical formulations of time dependent changes in ϵ and/or R_f may be introduced into the computer code by suitable substitution in the lines 14500 and/or 13700, respectively.

APPENDIX V: DIFMIG COMPARED TO ANALYTICAL SOLUTIONS

The validity of calculations performed using the program DIFMIG has been tested in the "single-barrier case", i.e. no barrier boundaries present, by comparison the DIFMIG solution to the analytical solution, which is obtainable in this particular case.

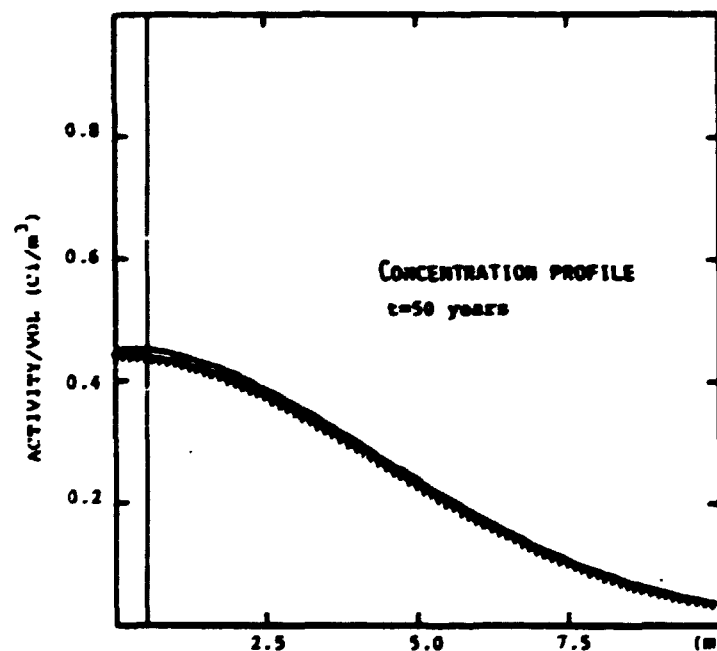
The analytical expression is given in equation (AV-1).⁸

$$C = \frac{1}{2}C_0 \{ \text{erf}[(h-x)/2\sqrt{Dt}] - \text{erf}[(h+x)/2\sqrt{Dt}] \} \quad (\text{AV-1})$$

Calculated concentration and flux profiles are shown in figures AV-I and AV-II, respectively, where a good agreement between the DIFMIG solution and the analytical solution is noted.

Within the first 0.5 m of the column the initial concentration of a nuclide is assumed to be 1 Ci/m^3 ; elsewhere the initial concentration is equal to zero. In figure AV-I the concentration profile to the time $t = 50$ years (covering the first 10 m of the

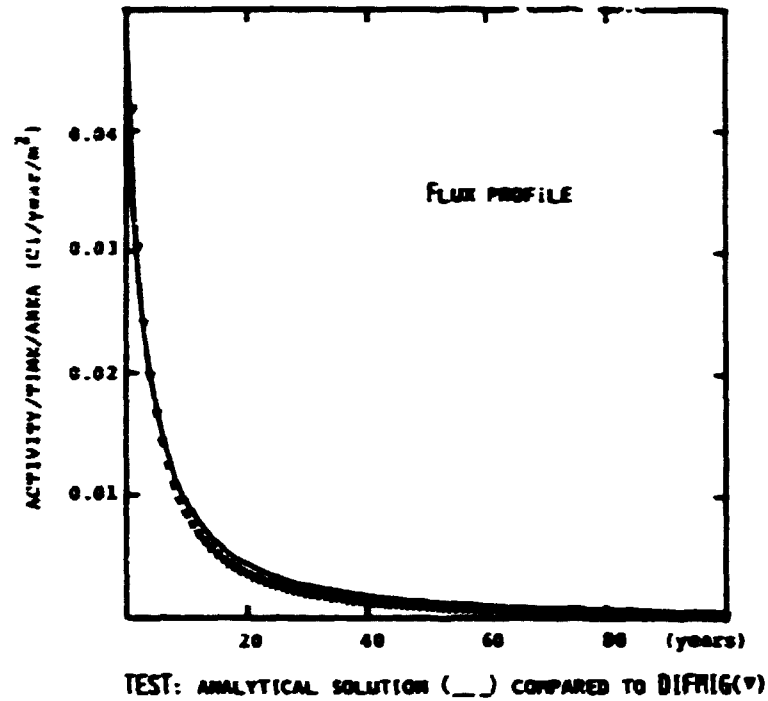
Figure AV-I



TEST: ANALYTICAL SOLUTION (—) COMPARED TO DIFMIG (•)

column) is shown. The flux out of the active core as a function of time is visualized in figure AV-II. For simplicity the decay constant for the nuclide is fixed equal to zero.

Figure AV-II



APPENDIX VI: EXAMPLES

Two types of barrier systems are considered: A) 4 barriers, the last barrier being of infinite length (figure AVI-I) and B) 4 barriers, the last barrier (of 1 m length) ending in a pond (figure AVI-II).

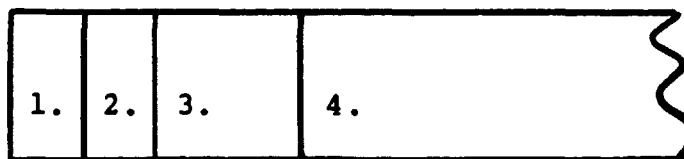


Figure AVI-I. 4-barrier system, the last barrier (4) being of infinite length

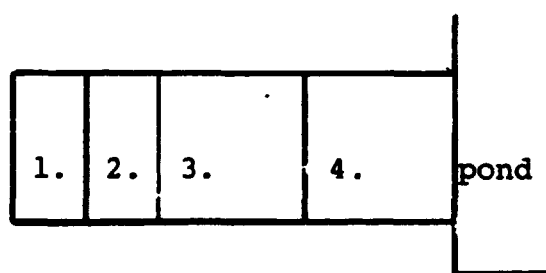


Figure AVI-II. 4-barrier system, the last barrier (4) ending in a pond.

The barrier 1 is assumed to be the active core of the repository, in which the nuclide under investigation is found at the time $t=0$ or produced as a function of time.

In the present examples the diffusion coefficient for the diffusing material is fixed equally in all barriers to $0.03 \text{ m}^2 \text{ year}^{-1}$ corresponding to the diffusion coefficient in pure water, the porosity is fixed to 0.4 (all barriers; except ex-iv), and the retention factors are 10^{-2} (barrier 1 and 2), 10^{-3} (barrier 3), and 10^{-2} (barrier 4), respectively.

ex-1: Barrier system A. At the time $t=0$ the concentration of a nuclide ($\lambda=0.001$) is 2000 Ci/m^3 in the active core (barrier 1)

(aqueous phase). No additional activity is produced during the period of time (2000 years) studied. In figure AVI-III the concentration profiles for $t=200, 400, 600, 800,$ and 1000 years, respectively, is shown (only the barriers 1, 2, and 3 are visualized). In figure AVI-IV the corresponding flux profiles for the fluxes of activity out from the barriers 1 and 2 are depicted.

Figure AVI-III

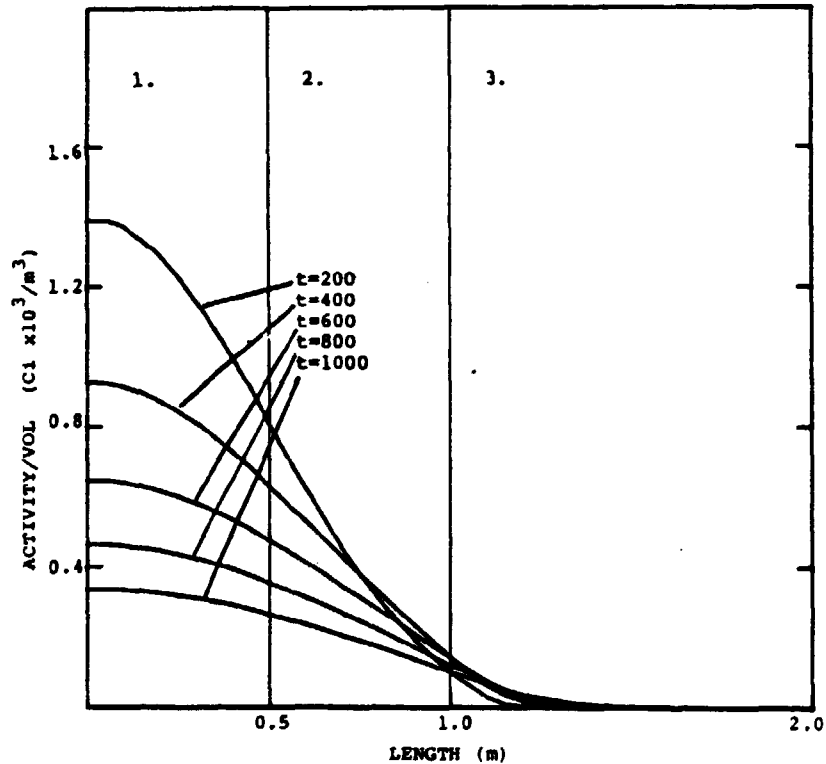
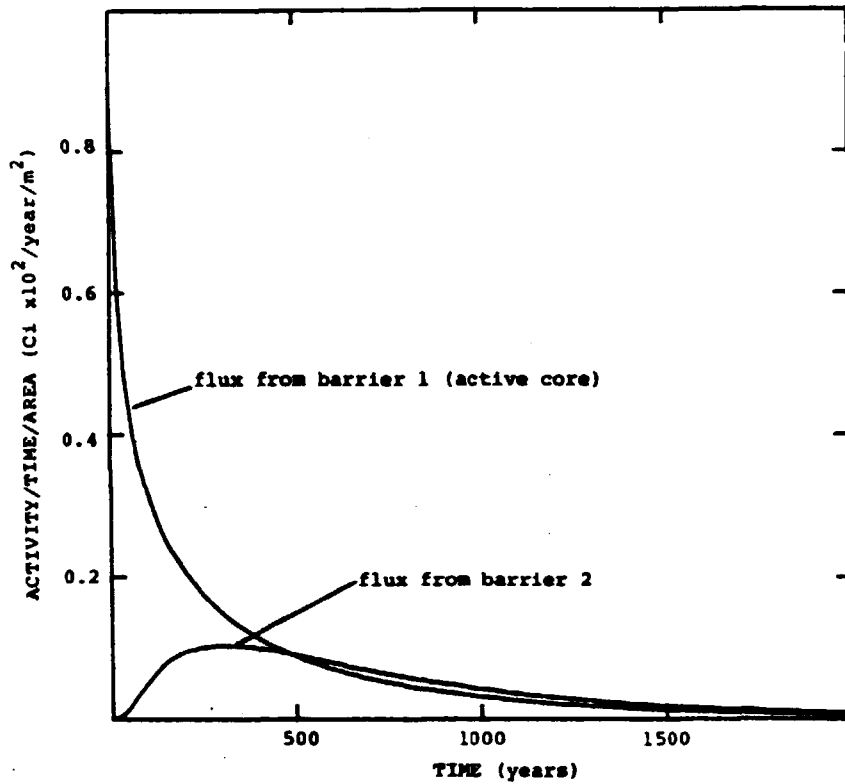


Figure AVI-IV



ex-ii: Barrier system A. At the time $t=0$ the concentration of the nuclide ($\lambda=0.001$) in the active core (barrier 1) is equal to zero. Activity is continuously supplied to the aqueous phase (1 Ci/year) during the time period studied (2000 years). In figure AVI-V the concentration profiles (through barriers 1-3) for $t=200, 400, 600, 800$, and 1000 years are shown. The fluxes of activity out from barrier 1 and 2 are visualized in figure AVI-VI.

Figure AVI-V

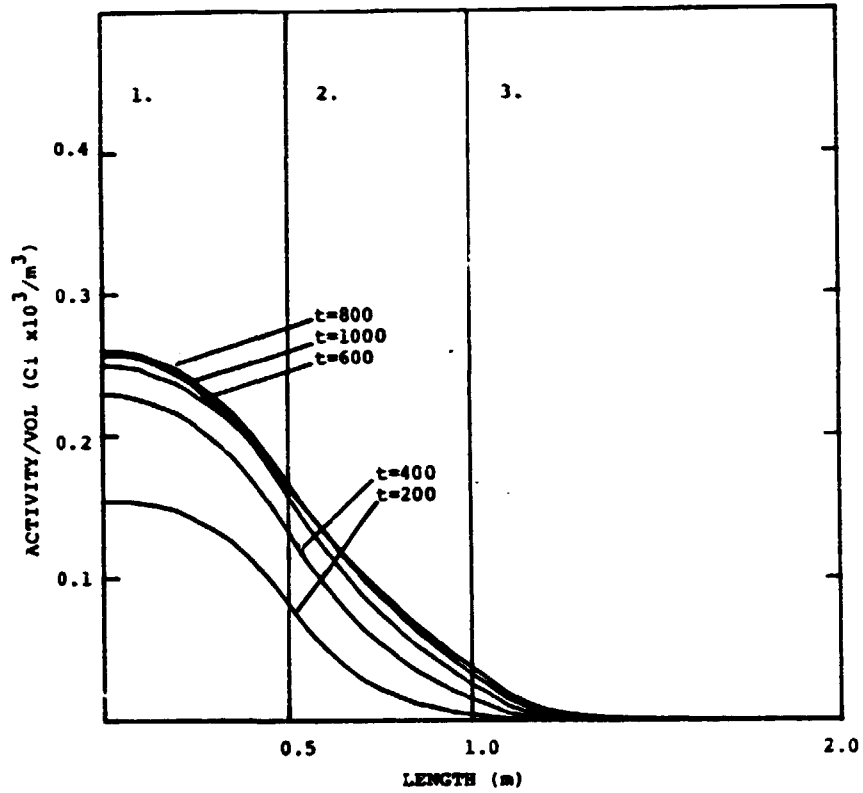
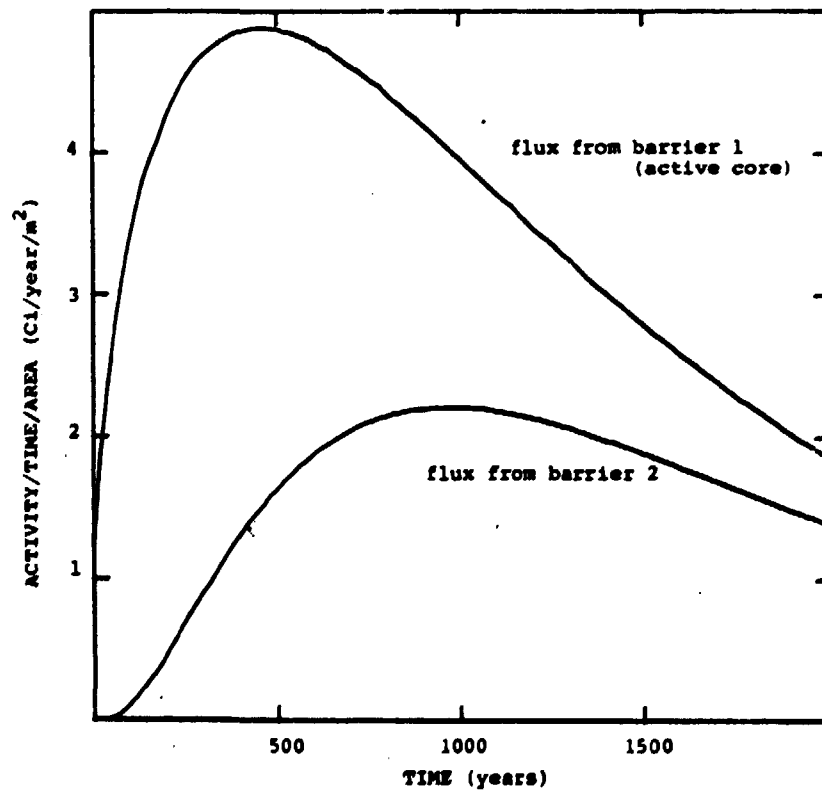


Figure AVI-VI



ex-iii: Barrier system B. At the time $t=0$ the concentration of a nuclides in the active core (barrier 1) is assumed to be 2000 Ci/m^3 (aqueous phase). No additional activity is produced during the period of time under investigation (2000 years).

Assuming a volume of the pond equal to 1 m^3 and a pump out rate equal to $1 \text{ m}^3/\text{year}$, the resulting variation of activity in the pond as a function of time is visualized in figures AVI-VII and figure AVI-VIII for two different nuclides with decay constants equal to 0.01 and 0.001, respectively.

Figure AVI-VII

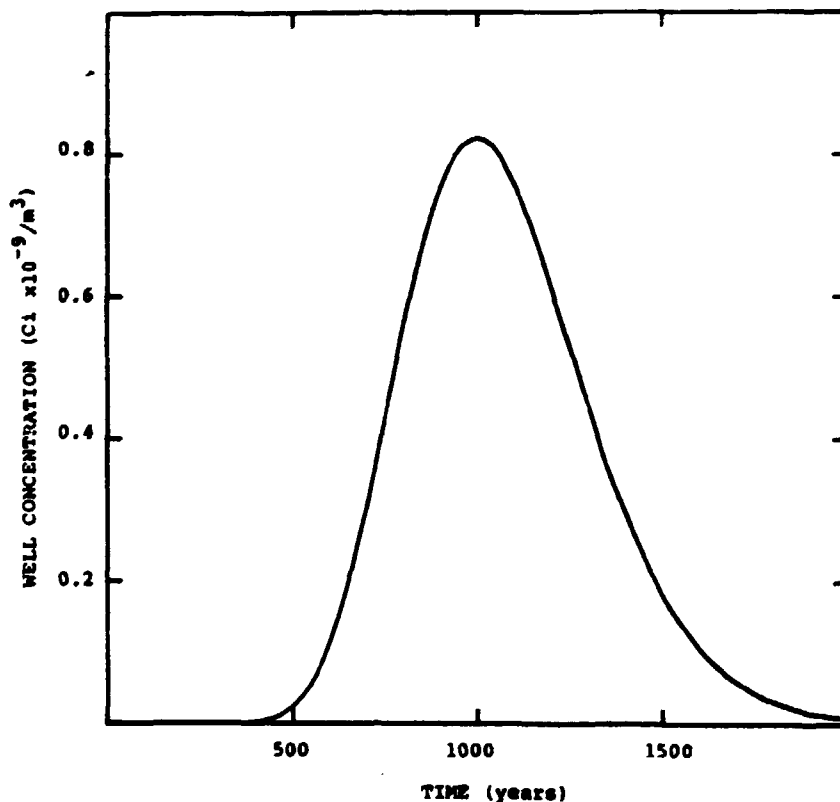
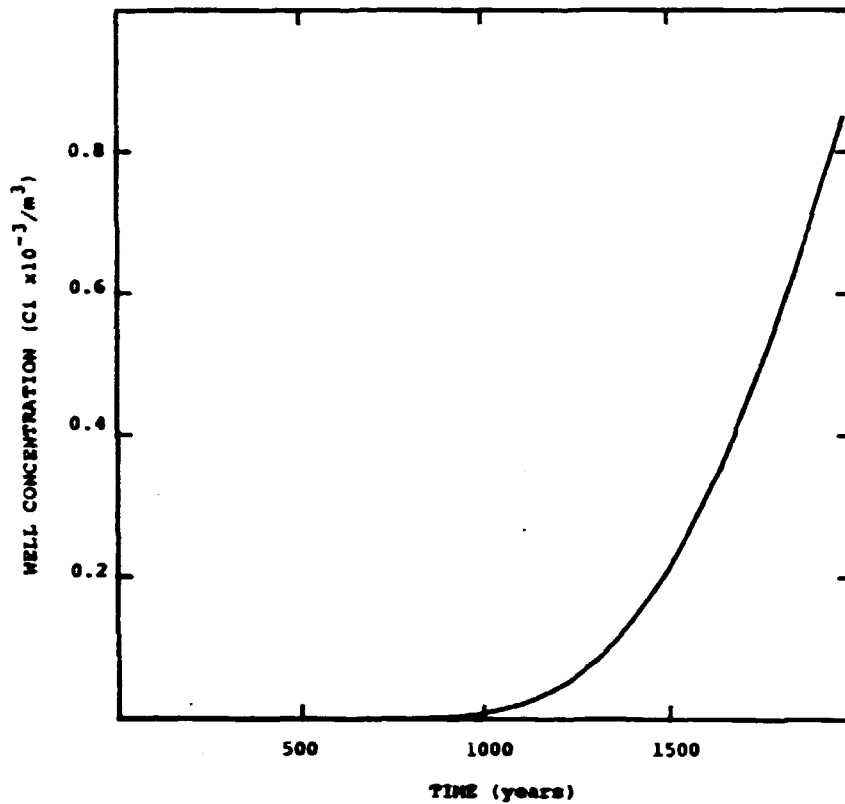
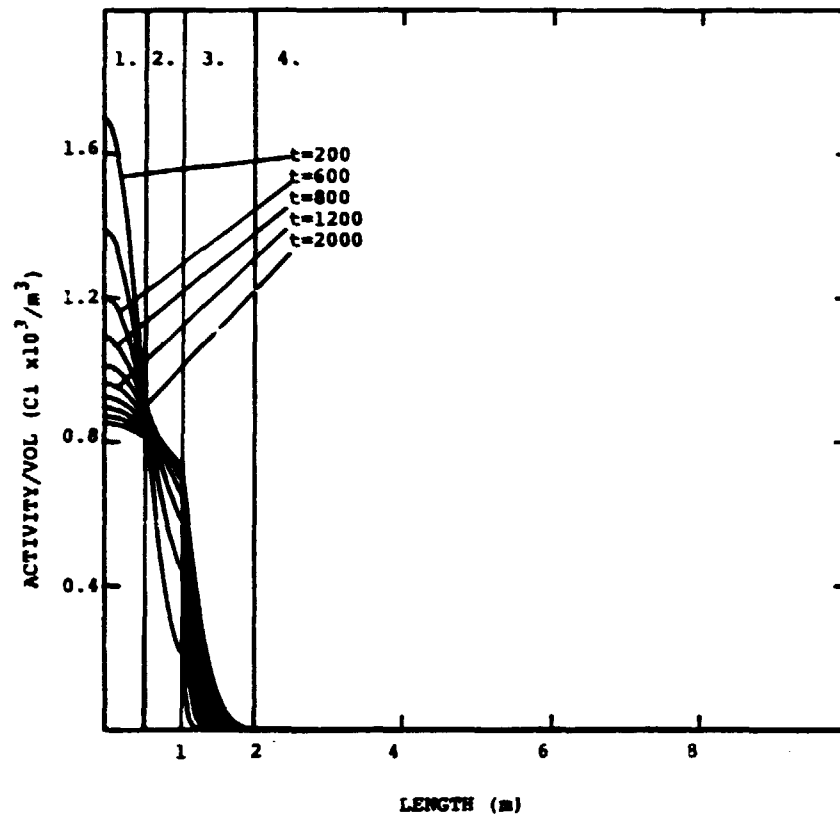


Figure AVI-VIII



ex-iv: Barrier system A. As ex-i, however, with the porosity in the barriers 1 and 2 equal to 0.4, in the barrier 3 equal to 0.04, and in barrier 4 equal to 0.4. Figure AVI-IX depicts the concentration profiles through the barriers for $t=200, 400, 600, 800, 1000, 1200, 1400, 1600, 1800$, and 2000 years, respectively.

Figure AVI-IX



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<p>Title and author(s)</p> <p>DIFMIG - A computer program for calculation of diffusive migration through multi-barrier systems</p> <p>Peter Bo and Lars Carlsen</p>	<p>Date November 1981</p> <p>Department or group CHEMISTRY</p> <p>Group's own registration number(s)</p>
<p>32 pages + tables + illustrations</p>	
<p>Abstract</p> <p>The FORTRAN IV program DIFMIG calculates one-dimensionally (<u>i.e.</u> column) the diffusive migration of single substances through arbitrary multibarrier systems according to the diffusion equation</p> $\partial C / \partial t = D'(t) \partial^2 C / \partial x^2 + F'(C, t)$ <p>where $D'(t)$ is the effective dispersion coefficient and $F'(C, t)$ is a function responsible for time dependent changes in concentration other than dispersion/diffusion (<u>e.g.</u> slow dissolution of a compound from a repository, radioactive decay, and/or build up of daughter products. The method takes possible time dependent variations in the effective dispersion into account.</p> <p>The diffusion equation is solved by a finite difference implicate method, the resulting trigonal matrix equation being solved by standard methods.</p> <p>Available on request from Risø Library, Risø National Laboratory (Risø Bibliotek), Forsøgsanlæg Risø), DK-4000 Roskilde, Denmark Telephone: (03) 37 12 12, ext. 2262. Telex: 43116</p>	<p>Copies to</p>